NEW DETAILS OF FERROELECTRIC SWITCHING

All of our current information technology relies on devices that process information as binary ones and zeroes. Ferroelectric materials are of special interest to developers of the next generation of such devices because they exhibit polarized electronic states that can represent bits of information. Moreover, these materials retain their polarization states without consuming electrical power, making ferroelectrics the subject of intense study for nonvolatile memory applications in which data is stored even when the power is turned off. One problem, however, is polarization fatigue: after a number of cycles, the switchable polarization begins to taper off, rendering the device unusable. Recently, a team of researchers from the University of Wisconsin, Bell Laboratories, and the University of Michigan used the x-ray synchrotron at the APS to study the micron-scale details of polarization fatigue in ferroelectric oxides.

"X-rays penetrate right through materials, so we can look deep inside electronic devices," says Eric Isaacs, former Bell Labs researcher, now director of Argonne's Center for Nanoscale Materials and a collaborator in the research. "Here we are studying materials in a realistic structure, not contrived devices suited to a particular probe."

Polarization fatigue is a well known problem in ferroelectric capacitor technology. In this case, the effect is believed to stem from migration of oxygen atoms to the electrode region. This, in turn, leads to formation of oxygen vacancies which can pin polarization domain walls and inhibit switching. Another mechanism involves the formation of a layer near the electrode interface that reduces the total electric field in the ferroelectric material and shuts down its ability to reverse polarization. Either way, the proposed mechanisms appear to be microscopic in nature, and so to study them in detail a precise high-resolution structural analysis tool is required. X-ray microdiffraction can image the evolution of polarization domains in buried ferroelectric thin films during switching with submicrometer resolution.

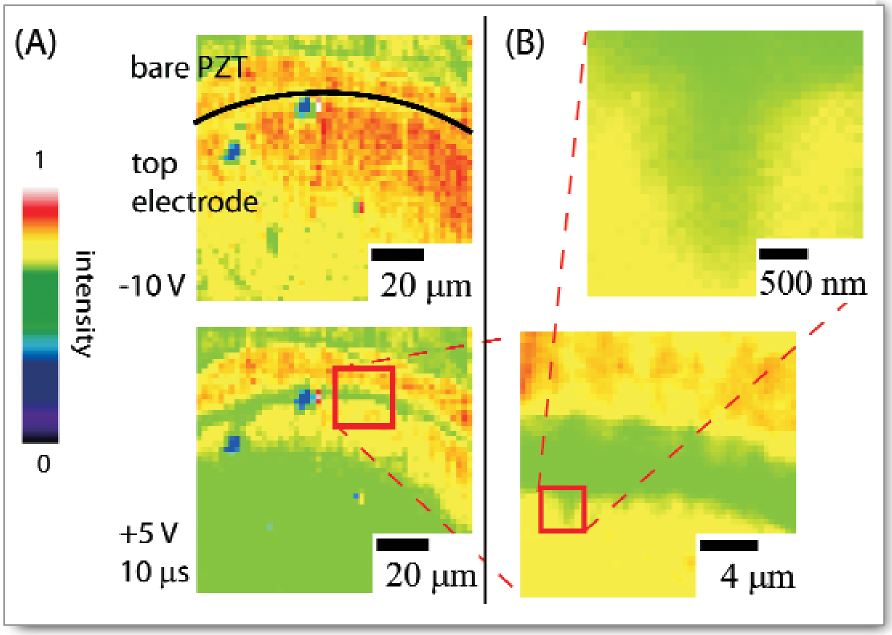

Fig. 1. Microdiffraction images of polarization switching in a $Pb(Zr,Ti)O_3$ thin film capacitor. (A) Images made following a -10 V pulse (top) and a shorter 10 µs +5 V pulse (bottom) show nearly complete polarization switching. Longer electrical pulses completely switch the device. (B) The boundary between two regions with opposite polarizations.

The ferroelectric devices were made in the University of Wisconsin group of Chang-Beom Eom by first depositing an $SrRuO_3$ bottom electrode on an insulating $SrTiO_3$ substrate. Epitaxial $PbZr_{1-x}Ti_xO_3$ (PZT) films with a nominal composition x = 0.55 and thicknesses of 80 or 160 nm were grown on top of the electrodes by radio frequency sputtering, followed by a top electrode layer consisting of sputtered polycrystalline platinum. A beam of 10 keV x-rays from the MHATT/XOR facility at beamline 7-ID of the APS were focused onto a 0.8-µm spot on this ferroelectric structure, and the diffracted x-rays were detected using conventional x-ray diffraction techniques. Images of the stored polarization with the ferroelectric layer were obtained by scanning the x-ray beam across the device. An advantage of this configuration is that electrical measurements can be made *in situ* during x-ray diffraction experiments. As a baseline, diffraction images were collected for each of the two stable polarization states, which established that x-ray microdiffraction was an accurate probe of the ferroelectric behavior.

The results of this study showed that polarization fatigue was qualitatively different when the switching was driven by lower-amplitude electric-field pulses (0.625 MV per cm peak) switching versus higher field pulses (1.2 MV per cm peak). Fatigue was observed in both regimes as the polarization-field hysteresis loops collapsed after repeated cycling with triangle wave pulses at 1 kHz. Low electric field fatigue was observed within 10^4 pulses as the PZT layer structure became pinned into an unswitchable state, which could be restored by exposure to higher field pulses.

A different process was found for fatigue induced by high field pulses. Although the onset of fatigue occurred after a much higher number of electric field cycles, the decrease in switchable polarization and the structural changes were more dramatic and irreversible. The x-ray microdiffraction images showed that isolated regions of severely decreased x-ray scattering intensity begin to form and that these eventually coalesce to encompass the entire region under the electrodes. The diffraction data indicate that there is a drastic loss of structural order as the fatigue progresses to failure of the device. "These are not things you can tell just from an electrical measurement," says collaborator Paul Evans of the University of Wisconsin. "There seem to be two sets of mechanisms in the low-field versus the high-field fatigue, and we can distinguish them by probing the structure with x-rays."

These measurements, made possible by the high brightness of the third generation synchrotron at the Advanced Photon Source, confirm that several mechanisms may be at play during fatigue and failure of ferroelectric devices. According to Evans, the results also indicate that x-ray microdiffraction is an ideal tool for high resolution studies of structural changes in thin film devices under a wide range of conditions, especially when structural and electronic phenomena are deeply enmeshed. "Microdiffraction is one of the killer applications for third generation synchrotrons such as APS."

— David Voss

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